## Supporting Information for

## Two New Layered Metal Chalcogenide Frameworks as Photocatalysts for Highly Efficient and Selective Dye Degradation

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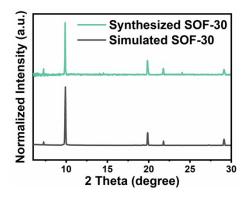


Figure S1. Powder X-ray diffraction (PXRD) patterns of as-synthesized SOF-30 and its simulated one.

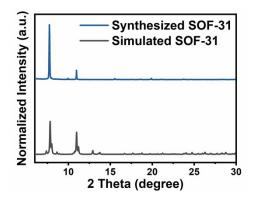


Figure S2. Powder X-ray diffraction (PXRD) patterns of as-synthesized SOF-31 and its simulated one.

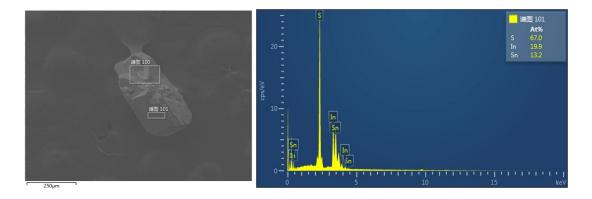


Figure S3. Left: SEM image of as-synthesized SOF-30. Right: EDS of SOF-30.

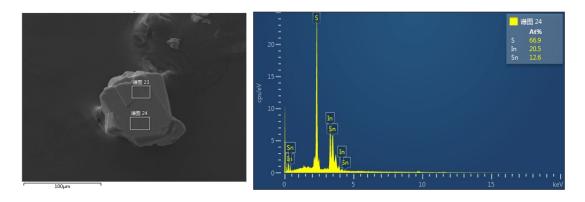


Figure S4. Left: SEM image of as-synthesized SOF-31. Right: EDS of SOF-31.

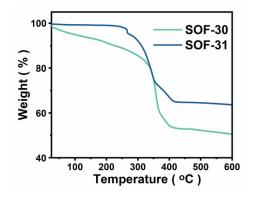


Figure S5. TG curves of SOF-30 and SOF-31. The initial gradual weight loss of 15% and 8% between 30-100°C could be attributed to loss of moisture and solvent adsorbed on the surface of SOF-30 and SOF-31. A further abrupt weight loss of 47% and 35% between 200-450°C are attributed to the carbonization of template of SOF-30 and SOF-31, respectively.

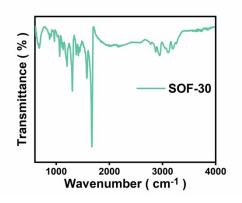


Figure S6. FT-IR spectrum of SOF-30.

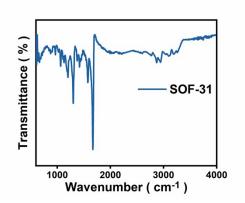


Figure S7. FT-IR spectrum of SOF-31.

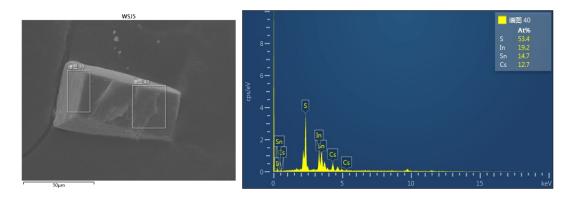


Figure S8. Left: SEM image of as-synthesized Cs@SOF-30. Right: EDS of Cs@SOF-30.

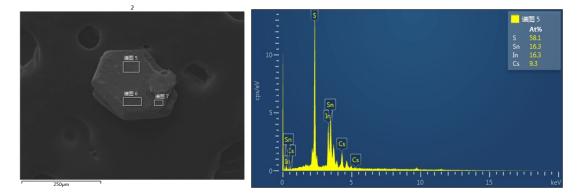


Figure S9. Left: SEM image of as-synthesized Cs@SOF-31. Right: EDS of Cs@SOF-31.

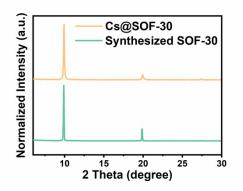


Figure S10. Powder X-ray diffraction (PXRD) patterns of Cs@SOF-30.

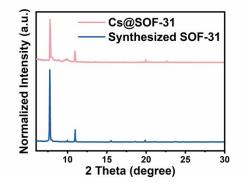


Figure S11. Powder X-ray diffraction (PXRD) patterns of Cs@SOF-31.

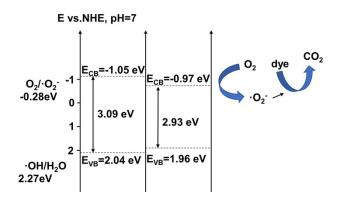


Figure S12. Schematic representation of the band-edge positions of in SOF-30 and SOF-31.

Let take **SOF-30** as example. The minimum of CB in **SOF-30** is negative than  $E_0^{2/.0^2}$ -(-0.28 V vs. NHE, pH=7). When the electrons are excited from the valence band (VB) to the CB under light irradiation, the photogenerated electrons can capture surface chemically adsorbed O<sub>2</sub> molecules to yield the superoxide radical  $\cdot$ O<sub>2</sub><sup>-</sup>, which could further participate in the degradation of dye molecules. In addition, the VB is positive than  $E_{.OH/H20}$  (+2.27 V vs. NHE, pH=7), the holes in VB can oxidize H<sub>2</sub>O molecules to yield reactive  $\cdot$ OH species. These reactive radicals contribute to the degradation of dye to H<sub>2</sub>O and CO<sub>2</sub> as the final products.

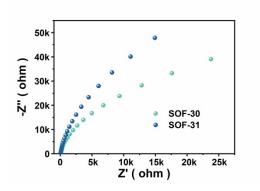


Figure S13. EIS Nyquist plots of SOF-30 and SOF-31 electrodes.

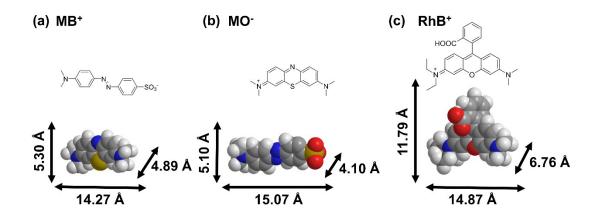
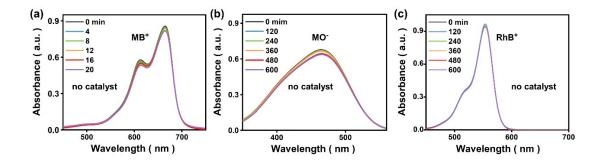
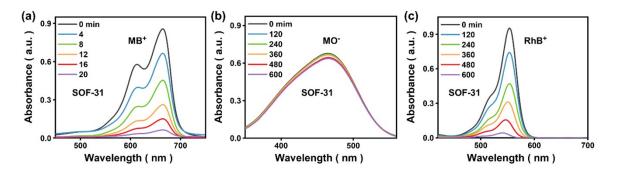


Figure S14. Chemical structures and dimensions (Å) of different dye molecules.



**Figure S15.** Absorbance under the photocatalytic degradation conditions without catalyst for  $MB^+(a)$ ,  $MO^-(b)$  and  $RhB^+(c)$ .



**Figure S16.** Absorbance under the photocatalytic degradation conditions of **SOF-31** for  $MB^+(a)$ ,  $MO^-(b)$  and  $RhB^+(c)$ .

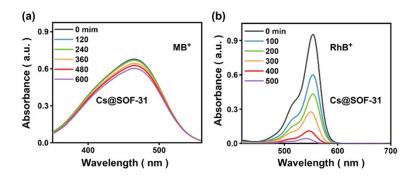


Figure S17. Absorbance under the photocatalytic degradation conditions of Cs@SOF-31 for MB<sup>+</sup> (a)and RhB<sup>+</sup>(b).

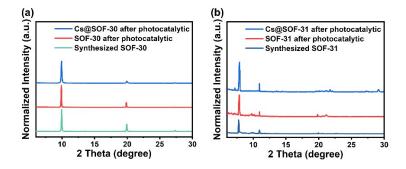


Figure S18. PXRD patterns of as-synthesized SOF-30 and Cs@SOF-30 (a); SOF-31 and Cs@SOF-31 (b) after photocatalytic experiment.

Compounds	SOF-30	SOF-31
Framework formula	$[In_4Sn_2S_{12}]$	$[In_5Sn_3S_{17}]$
Formula weight	1081.38	1494.54
Crystal system	Monoclinic	Orthorhombic
Ζ	4	4
Space group	$P2_{1}/c$	Pccn
<i>a</i> (Å)	13.0435 (7)	23.9507 (16)
<i>b</i> (Å)	35.1784 (17)	12.4142 (9)
<i>c</i> (Å)	10.3323 (5)	22.5931 (16)
α (°)	90	90
$\beta$ (°)	91.933 (2)	90
γ (°)	90	90
$V(Å^3)$	4738.3 (4)	6717.6 (8)
<i>T</i> (K)	119.89	119.99
<i>F</i> (000)	1952.0	2688.0
$D(g/cm^{-3})$	1.516	1.478
Collected reflections	46448	34257
Independent reflections	8658	6452
GOF on F <sup>2</sup>	1.042	1.008
$R_1, wR_2(I \ge 2\sigma(I))$	0.0423,0.1095	0.0493,0.1390
$R_1$ , $wR_2$ (all data)	0.0580,0.1189	0.1289,0.1906

 Table S1. Crystal data and refinement results of SOF-30 and SOF-31.

 Table S2. Elemental analysis results of SOF-30 and SOF-31.

	Elements (wt.)	N (%)	C (%)	H (%)
SOF-30	Calculated	6.35	20.08	3.20
	Experimental	6.42	19.78	3.20
SOF-31	Calculated	5.53	20.91	3.92
	Experimental	5.47	20.78	3.90